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THEORY OF THE ELEMENTARY PROCESSES

IN THE SPECTROPHONE

By

J. C. DECLUS

Department of Chamistry Oregon State College Corvallis, Oregon

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## I. THEORY OF THE LLEAFFMARY PROCESSES IN

#### THE SPECTROPHOME

#### A. INTRODUCTION

The use of the infrared spectrophone for the determination of the lifetime of vibrationally excited molecules was first described by Slobodskaya (1). This instrument consists essentially of a cell containing an infrared absorbing gas; the cell is illuminated intermittently with radiation corresponding to an absorption band of the gas and the periodic variations in pressure due to alternate heating and cooling are detected with a sensitive microphone. Changes in phase shift of this signal, relative to the phase of the radiation, are related to the mean vibrational lifetime of the excited molecules. The experiments of Slobodskaya were performed with various mixtures of  $CO_2$  and air, and results were reported for both infrared fundamentals  $(\omega_2 = 667 \text{ cm}^{-1} \text{ and } \omega_1 = 2350 \text{ cm}^{-1})$  as well as a combination band.

An analysis of this experiment was described by Stepanov and Girin (2), who considered a two state model of the vibrational levels and assumed square wave excitation.

A detailed analysis of the inelastic collision process in which vibrational energy is converted into translational energy has been given by Schwartz, Slawsky, and Herzfeld (3), whose results indicate that the probability of this sort of process is a very sensitive function of both the energy of the vibrational quantum and of the reduced mass of the collision. At room temperature and atmospheric pressure, the relaxation time for this process may vary from the order of the 10-6 sec. for vibrational modes of relatively low frequency, to as high as 1 sec. for

figure have not been observed for several reasons: in sound dispersion studies, for example, the equilibrium heat capacity due to such modes is too small to detect.

It is the purpose of the remainder of this paper to carry out an analysis of the rates of the elementary processes occurring in the spectrophone with emphasis upon the fact that the large range of the parameters, especially their sensitivity to the vibrational frequency, leads to complications in the interpretation of the observed phase shifts.

B. RATE OF CONVERSION OF RADIANT TO TRANSLATION EMERGY
It has been shown by Schwartz, Slawsky, and Herzfeld (3) that the
net rate of conversion translational into vibrational energy is

$$\frac{dE_{\mathbf{v}}}{dt} = \frac{1}{7} \left[ E_{\mathbf{v}}(T) - E_{\mathbf{v}} \right] \tag{1}$$

where 7 is a characteristic time constant measuring the life-expectancy of the excited vibrational state before its termination by an inelastic collision.  $E_{v}$  is the actual vibrational energy and  $E_{v}(T)$  is the vibrational energy which would be in equilibrium with the translational energy at translational temperature,  $T_{v}$ 

The net rate of absorption of photons must now be considered. For this purpose, the Einstein coefficients are required, namely  $A_{v \rightarrow v-1}$ ,  $B_{v \rightarrow v-1}$ , and  $B_{v \rightarrow v+1}$ , which are respectively the coefficients of spontaneous emission, induced emission, and induced absorption. Only the fundamental transitions involving unit change in the vibrational quantum number, v, are considered. These quantities are given by the following expressions,

$$A_{v} = v-1 = \frac{8\pi^2 \sqrt{2}}{3c^3} (\frac{3\mu}{30})^2 = \alpha v$$
 (2a)

$$B_{v \to v-1} = \frac{\pi}{3h\nu} \left( \frac{\partial \mu}{\partial \Theta} \right)^{v} = \beta^{v}$$
 (2b)

$$B_{v \to v+1} = \frac{\pi}{3hy} \frac{\partial \mu}{\partial Q} (v+1) = \beta (v+1)$$
 (2c)

in the case of the harmonic oscillator. The dipole derivative with respect to the normal coordinate, Q, is evaluated at equilibrium and it is assumed that only linear terms in Q appear in the expansion of  $\mu$ . Now if the number of molecules per unit volume in the vibrational state  $\mathbf{v}$  is indicated by  $\mathbf{n}_{\mathbf{v}}$  and if  $\rho$  is the density of radiation of appropriate frequency, the net rate of increase of vibrational energy, per unit volume, at the expense of radiation is

$$\frac{dEv}{dt} = hy \sum_{v=0}^{\infty} \left[ -\left( \frac{A}{v \rightarrow v-1} + B \right) + \frac{B}{v \rightarrow v-1} \right) + \frac{B}{v \rightarrow v-1} \right] n_v$$

$$= hy \left( -\alpha \sum_{v=0}^{\infty} vn_v + \beta \rho \sum_{v=0}^{\infty} h_v \right)$$

$$= hy \left[ -\alpha \left( \frac{E}{hy} - \frac{\mu_1}{\rho} \right) + \beta \rho \mu_1 \right] = -\alpha E_v + hy \left( \frac{\alpha}{\rho} + \mu_2 \rho \mu_1 \right)$$
(3)

In (3), n is the total number of molecules per unit volume.

In the spectrophone experiments to be considered, the radiation density,  $\rho$  , is modulated with angular frequency,  $\alpha$ , so that

Although in some cases the wave form may be more nearly square than sinusoidal, the detecting device is usually sharply tuned to the fundamental modulation frequency, so that we shall be primarily concerned

with the second term in (4), and will only use the first two terms in the following development.

By combining the expressions for interconversion of translational and vibrational energy and for interconversion of vibrational and radiant energy, using the modulation frequency,  $\omega$ , one finds

$$\frac{dE_{V}}{dt} = -(\alpha + \frac{1}{C})E_{V} + \frac{1}{C}E_{V}(T) + a_{o} + a_{s} \sin \omega t$$
 (5)

where 
$$a_0 = nh \mathcal{V} \left( \frac{N}{2} + \beta \mathcal{F}_0 \right)$$
 (5a)

$$a_1 = nh \mathcal{V} \beta f$$
 (5b)

If one now considers the gross heat balance in the spectrophone cell, and allows for heat loss only by conduction, the heat flow equation becomes

$$\mathbf{K} \nabla^2 \mathbf{T} - \mathbf{c} \sqrt{\frac{\partial \mathbf{I}}{\partial \mathbf{p}}} + \frac{1}{c} \left[ \mathbf{E}_{\mathbf{V}} - \mathbf{E}_{\mathbf{V}}(\mathbf{I}) \right] = 0 \tag{6}$$

in which K is the thermal conductivity,  $c_V$  the heat capacity (for unit volume), and the last term represents the rate of production of sensible heat by conversion of vibrational into translation energy. The simultaneous solution of (5) and (6) is considerably simplified in case  $h_V >> V T$ , since  $E_V$  (T) reduces to the zero point energy,  $h_V >> V T$ , i.e., a constant independent of temperature and hence of the time. Subject to such an assumption, the steady-state solution of (5) is given by:

$$E_{\mathbf{v}} = b_0 + b_1 \sin(\omega t + \phi) \tag{7}$$

with 
$$b_0 = \frac{\cosh \sqrt{\left[\frac{1}{2}(\alpha z + i) + (z e_0)\right]}}{\alpha z + 1}$$
 (73)

$$b_1 = \frac{\ln \sqrt{\beta \tau \gamma_1}}{\left[(\omega \tau)^2 + (\alpha \tau + 1)^2\right]^{1/2}}$$
(7b)

$$= \tan^{-1} \left\{ -\frac{\omega \gamma}{\chi \gamma + 1} \right\} \tag{7c}$$

Before proceeding to a discussion of the heat flow equation, we must consider the case in which  $E_v$  (T) cannot be put equal to  $nh\gamma/2$ . Since the actual variations of T are small,

$$E_{v}(T) = c_{v}! \Delta T + E_{v}(T_{o})$$

where  $c_v$  is the vibrational heat capacity per unit volume and at constant volume, and  $T = T_o + \triangle T$ . Equations (5) and (6) are replaced by

$$\frac{dEv}{dt} = -(\Omega + \frac{1}{2}) E_v + \frac{c_v!}{2} \Delta^T + \frac{E_v}{2} (To) + a_o + a_1 \sin \omega t$$
 (5')

$$\mathbf{K} \nabla^{2} (\Delta \mathbf{T}) - \mathbf{c}_{\mathbf{V}} \underline{\partial \Delta T} + \underline{\mathbf{E}_{\mathbf{V}}} - \underline{\mathbf{c}_{\mathbf{V}}} \Delta \mathbf{T} - \underline{\mathbf{E}_{\mathbf{v}}} (\mathbf{T}_{\mathbf{0}}) = 0$$
 (61)

A formal solution of (5') is obtained by putting

$$Ev = b_0! + b_1 \sin(wt + \phi) + \frac{c_y!}{7} \int \Lambda T dt$$

with 
$$b_0' = \frac{E_{\mathbf{v}}(T_0) + nh \gamma J(\frac{\alpha \gamma}{2} + \beta \gamma \beta_0)}{\alpha \gamma + 1}$$

#### C. SOLUTION OF THE HEAT FLOW EQUATION

First consider the case in which  $E_v$  (T) is a constant, namely  $nh\sqrt{2}$ .

The heat conduction equation (6) then becomes

$$K \nabla^{2} T - c_{V} \frac{\partial T}{\partial t} + \frac{b_{0}}{2} + \frac{b_{1}}{2} \sin(\omega t + \phi) = 0$$
 (8)

ii.

with the aid of (7). Again, we are interested in the steady-state, periodic solution of (8). Although various boundary values may be assumed, we shall consider only the simple one-dimensional problem, T = Tx, the with T(o,t) = T ( $\ell$ ,then  $\ell$ ) representing the length of the cell. The assumption of a solution of the form

$$T(x,t) = f(x) + g(x) \sin(\alpha x + \phi) + h(x) \cos(\alpha x + \phi)$$
 (29)  
together with the above boundary conditions leads to rather clumsy expressions for g and h involving linear combinations of the functions  $\sin x \sin x$ ,  $\sinh x \cos x$ , and  $\cosh x \sin x$ , where

$$\lambda^2 = \frac{2K}{\omega c_v} \tag{10}$$

The average values of g(x) and h(x) over the length of the cell, simplify somewhat to yield the following expressions:

$$\overline{g} = \frac{-b_1}{\omega \overline{c_v}} \left(\frac{\lambda}{\rho}\right) \left[ \frac{\sinh \frac{\ell}{\lambda} \cos \frac{\ell}{\lambda} + \cosh \frac{\ell}{\lambda} + \sin \frac{\ell}{\lambda} - \sinh \frac{\ell}{\lambda} \cosh \frac{\ell}{\lambda} - \sinh \frac{\ell}{\lambda} \cos \frac{\ell}{\lambda}}{\sinh^2 \frac{\ell}{\lambda} + \sin^2 \frac{\ell}{\lambda}} \right] \\
= \frac{b_1}{\omega \overline{c_v}} \left(\frac{\lambda}{\ell}\right) \left[ \frac{\sinh \frac{\ell}{\lambda} \cosh \frac{\ell}{\lambda} - \sinh \frac{\ell}{\lambda} \cos \frac{\ell}{\lambda} + \cosh \frac{\ell}{\lambda} \sin \frac{\ell}{\lambda} - \sinh \frac{\ell}{\lambda} \cos \frac{\ell}{\lambda}}{\sinh^2 \frac{\ell}{\lambda} + \sin^2 \frac{\ell}{\lambda}} \right] \\
= \frac{b_1}{\omega \overline{c_v}} \left(\frac{\lambda}{\ell}\right) \left[ \frac{\sinh \frac{\ell}{\lambda} \cosh \frac{\ell}{\lambda} - \sinh \frac{\ell}{\lambda} \cos \frac{\ell}{\lambda} + \sinh \frac{\ell}{\lambda} \cos \frac{\ell}{\lambda} + \sinh^2 \frac{\ell}{\lambda}}{\sinh^2 \frac{\ell}{\lambda} + \sinh^2 \frac{\ell}{\lambda}} \right]$$
(12)

If the condition  $\langle \rangle \rangle \lambda$  obtains, the expressions for  $\overline{g}$  and  $\overline{h}$  simplify to  $\frac{\overline{g}}{a^{b_1}c^{c_v}} \frac{\lambda}{\ell}$ ,  $\frac{\overline{h}}{a^{b_1}c^{c_v}} \frac{b_1}{\ell}$ ,  $\frac{b_1}{a^{b_1}c^{c_v}} \frac{\lambda}{\ell}$ ,  $\frac{\overline{h}}{a^{b_1}c^{c_v}} \frac{b_1}{\ell}$ 

whence it follows that the time dependent part of T, averaged over the cell length, is

$$T = \frac{b_1}{\omega z c_v} \left[ \frac{\lambda}{\ell} \sin \left( \omega t + \phi \right) + \left( \frac{\lambda}{\ell} - 1 \right) \cos \left( \omega t + \phi \right) \right]$$
 (13)

which may finally be written in the form

$$T = b_1 \left(1 - 2\lambda + 2\lambda^2 \right)^{1/2} \sin(\omega t + \phi + \phi^2)$$

$$\frac{1}{\omega} C_{\nu}$$
(14)

$$\phi = \tan^{-1}\left(1 - \frac{\ell}{\lambda}\right) \tag{15}$$

It is apparent that when  $l \gg \lambda$ , will be very close to -N/2, and although  $\lambda$  may vary slightly if the pressure or composition of the gas in the spectrophone cell is altered,  $\phi$  will remain almost constant.

Turning to the case in which  $E_{\mathbf{v}}(T)$  is not constant, we shall neglect the dependence of phase shift upon heat conduction, which is simply equivalent to letting  $K \rightarrow 0$  in (8). In such a case, the functions g and h assume the values:

$$g = \frac{1}{2} \frac{\chi^{2} \gamma^{b_{1}}}{c \sqrt{(\gamma - \omega^{2} \chi^{2})^{2} + \omega^{2} \chi^{2} \gamma^{2}}}$$
(16)

$$h = \frac{(\gamma - \omega^2 \gamma^2)\omega \gamma c_1}{c_v \left[ (\gamma - \omega^2 \gamma^2)^2 + \omega^2 \gamma^2 \gamma^2 \right]}$$
(17)

in which =  $c_v^{1/c_v}$ , i.e., the ratio of the vibrational heat capacity to

the translational heat capacity. Here the phase shift turns out to be

$$\phi'' = \tan^{-1} \left\{ \frac{\gamma - \omega^2 \tau^2}{\omega \tau \gamma} \right\}$$

Continuing the assumption that W is high enough so that heat conduction does not play an important role, we now combine the results of (7), (7b), (7c), (16), and (17) to obtain an expression which gives the temperature amplitude and the overall phase shift:

$$\Delta T = \Delta T_i \sin(\omega t + \phi)$$

and

which 
$$\Delta T_1 = \omega z n h \nu |z z r| [(y - \omega^2 z^2)^2 + (\omega z r)^2]^{1/2} [(\omega z)^2 + (x z + 1)^2]^{1/2}$$

$$= \frac{(\omega z^2 A^4 I_1 [(x - \omega^2 z^2)^2 + (\omega z r)^2]^{-1/2} [(\omega z)^2 + (x z + 1)^2]^{-1/2}}{c_V}$$

$$\phi = tan^{-1} \left[ \frac{-(\omega^2 z^2)^2 + (x z + 1)(x - (\omega^2 z^2)^2)}{\omega z [(x - \omega^2 z^2 + (x z + 1))^2]^{-1/2}} \right]$$
(20)

Here the integrated absorbtion coefficient, A\*, at the gas density employed, has been introduced; see Thorndike, Wells, and Wilson (5). I, is the intensity of the modulated radiation.

Some simplification of (20) is greatly desirable in order to relate the observed phase shift to  $\mathcal{T}$ . Other factors being equal, a small value of  $lpha \, \mathcal{T}$  in comparison with unity leads to a large amplitude and to the following simplification of (20). If  $\chi \gamma \ll 1$  and if  $\omega^2 c^2 < \gamma$ , (20) becomes approximately

since  $\omega_{\mathcal{T}}$  is then necessarily small compared with unity. This situation is realized experimentally for lower frequency molecular modes, say below 1000 cm<sup>-1</sup>, where  $\tilde{c}$  is less than  $10^{-4}$  sec,  $\gamma \approx 0.1$ , with appropriate choice of  $\omega$  .

For the higher frequency molecular modes, say above 2000 cm<sup>-1</sup>,  $\mathcal{T}$  may become quite large, perhaps  $10^{-2}$  seconds or greater. Using Eggers and Crawford's (4) value of  $\frac{\partial \mathcal{H}}{\partial Q}$  for the stretching mode of  $CO_2$ , Q  $\mathcal{T}$  would be 42.7 with  $\mathcal{T}=10^{-2}$  sec. Thus, for high frequency molecular modes of high intensity, we may perhaps need to consider the case in which  $(X\mathcal{T})$ , which simplifies  $\Phi$  to

provided  $\omega_{7<1}$ . This latter situation is relatively unfavorable, however, for the following reasons: (i) when  $\varepsilon$  is large, a very small (i) must be employed to justify the simplification of  $\tan^{-1}$  just employed; (ii) this leads to relatively large conduction heat loss, In any case, it appears that Slobodskaya's (1) identification of the observed time lag with  $\varepsilon$  is not justified for the 2350 cm<sup>-1</sup> band of  $\varepsilon_{0}$  when a modulation frequency of  $\omega = 1572$  sec.<sup>-1</sup> was employed.

## D. CONCLUSION

It has been shown that the phase shift of the temperature variations in the spectrophone cell, averaged over the length of the cell, depends in a complex manner upon the following variables:

X = probability of spontaneous emission

'Y = ratio of vibrational to translational heat capacity

(4) = modulation frequency

 $\mathcal{T}$  = mean lifetime for inelastic collisions.

Although  $(\chi)$  and  $\Upsilon$  are known with good accuracy for the modes of vibration to be studied experimentally, it appears necessary to work under conditions such that the observed phase shift is simply related to  $\mathcal{C}$ :

this situation is attained experimentally most easily for low frequency molecular modes, e.g., less than 1000 cm<sup>-1</sup>, or for higher frequencies in the case of collisions with very light molecules such as  $H_2$  or  $H_2$ . It is further necessary that  $\omega^2 \sim 2$ . Since  $\gamma$  is 0.133 when the molecular frequency is 1000 cm<sup>-1</sup>, and Tmay be of the order  $10^{-4}$ ,  $\omega$  should be less than  $10^2$ .

When Mz is much less than unity, and, in addition,  $\omega^2 z^2 < \gamma$  the amplitude of the temperature variations reduces to

$$\Delta T_1 = \frac{\omega z^2 A^* I_1}{\gamma c_V}$$

$$\Delta T_1 = \frac{A^* T_1}{\omega^2 c G v}$$

It is thus apparent that optimum response is obtain when  $\omega_{\mathcal{C}}$  is of the order of  $\gamma''^2$ , a condition which is somewhat in conflict with the optimum situation for a simple relation between  $\phi$  and  $\mathcal{C}$ . If  $\gamma$  vanishes, the response improves as  $\omega$  decreases, but this conclusion, of course, neglects the effect of heat conduction, which demands that

$$\omega \gg \frac{2\kappa}{\ell^2 c_V}$$

for good response.

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